

PATENT ABSTRACTS OF JAPAN

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(54) METHOD FOR SEPARATING AND PURIFYING CARBON NANOTUBE

(57)Abstract:

PURPOSE: To provide a method capable of relatively easily removing carbon impurities from the carbon nanotubes containing the carbon impurities and obtaining the carbon nanotubes in a high quality.

CONSTITUTION: The method for separating and purifying the carbon nanotubes comprises finely grinding carbon nanotubes containing carbon impurities into the fine product having an average particle diameter of $0.47\mu\text{m}$, dispersing the finely ground product in a liquid, centrifuging the dispersion, filtering the formed supernatant containing fine particles having an average particle diameter of $\leq 0.2\mu\text{m}$, and subsequently backing the filtration product at a high temperature.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] this invention relates to the method of carrying out separation refining of the carbon nanotube from the carbon nanotube containing a carbon impurity.

[0002]

[Description of the Prior Art] The physical properties attract attention as a form of the new carbon with which the carbon nanotube discovered recently follows fullerene, and application in the wide range field from a material science to electronics is expected. The method of compounding this carbon nanotube in large quantities is proposed by Nature 358 and 220 (1992).

[0003] After having carried out crushing of the carbon nanotube rough product, having immersed the crushing object into ethanol, since carbon impurities, such as graphite-like matter and amorphous carbon, were intermingled in large quantities, performing ultrasonic irradiation in the carbon nanotube rough product obtained by the method of the above-mentioned reference and making a carbon nanotube and a carbon impurity divide into it, separation refining of the carbon nanotube was carried out by collecting carbon nanotubes as a distributed object.

[0004] However, since the carbon impurity was incorporated in the aggregate at the time of condensation of the carbon nanotube which is the fibrous material which is easy to condense, it was difficult to separate the carbon impurity incorporated in this aggregate only by distributing a crushing object in ethanol. For this reason, in the above-mentioned carbon nanotube separation refining object, many carbon impurities, such as a graphite particle and amorphous carbon, were contained

[0005]

[Problem(s) to be Solved by the Invention] this invention removes this carbon impurity from the carbon nanotube containing a carbon impurity by comparatively easy operation in view of the actual condition of such conventional technology, and makes it the technical problem to offer the method of obtaining a quality carbon nanotube.

[0006]

[Means for Solving the Problem] This invention persons came to complete this invention, as a result of repeating examination wholeheartedly that the above-mentioned technical problem should be solved. That is, by according to this invention, carrying out pulverizing processing of the carbon nanotube containing a carbon impurity at 0.4 micrometers or less of pitch diameters, making it distribute in a liquid and carrying out centrifugal separation of the pulverizing object, the supernatant containing the particle of 0.2 micrometers or less of pitch diameters is formed, and the separation refining method of the carbon nanotube which consists of carrying out elevated-temperature baking of the distributed object which filtered and obtained this supernatant is offered.

[0007] this invention is explained in full detail below. What is set as the object of processing in this invention distributes in alcohol, such as ethanol, what carried out crushing of the carbon nanotube containing the above-mentioned carbon impurity, i.e., the carbon nanotube rough product, and collects the distributed objects. In this invention, pulverizing processing is first performed to the carbon nanotube containing this carbon impurity, and it considers as the pulverizing object of 0.4 micrometers or less of pitch diameters. About a carbon nanotube, since the diameter of a carbon nanotube is about dozens of nm, a pitch diameter shall express average length here. In the carbon nanotube containing the above-mentioned impurity, since many things in the state where the carbon nanotube adhered to the graphite-like matter can also be seen, in order to remove the graphite-like matter certainly, it is necessary to consider as the pulverizing object of 0.4 micrometers or less of mean particle diameters.

[0008] Although dry type processing is also possible for the pulverizing processing performed here, a wet process is used preferably. By this wet pulverizing processing, the carbon nanotube containing a carbon impurity is pulverized with the pulverizing means by which it is well-known for example, in alcohol. Ethanol, a methanol, a butanol, etc. can be used as the above-mentioned alcohol. The water which added the dispersant besides alcohol is usable. A pulverizing object is dried after pulverizing. This pulverizing object is a carbon nanotube, the graphite-like matter, and the mixture of amorphous carbon substantially.

[0009] Next, after distributing the dried pulverizing object in a liquid, centrifugal separation is carried out, a supernatant is formed, this supernatant is filtered, and a distributed object is obtained. As the above-mentioned liquid, what added the dispersant to pure water can be used, for example. In this case, as a dispersant, a sodium oleate, polyoxyethylene alkyl phenyl ether, a polyoxyethylene polyol carboxylate, etc. can be used. 0.05 - 0.3 % of the weight is suitable for the addition of a

dispersant to water. In distribution, you may perform ultrasonic irradiation. Centrifugal separation is centrifugal forces 8000-12000G, and it is desirable to carry out a grade for 30 - 60 minutes. By the above-mentioned centrifugal separation, since the graphite-like matter of a particle diameter 0.3 micrometers or more sediments, 70 - 80% of the graphite-like matter sediments, and the supernatant has become what graphite-like matter, remaining carbon nanotubes, and remaining amorphous carbon distributed as a particle of 0.2 micrometers or less of pitch diameters. Filtration of a supernatant can be performed using the micropore filter of an about 0.2-0.5-micrometer aperture etc. Most amorphous carbon is removed by this filtration. The filtered distributed object is dried.

[0010] Next, elevated-temperature baking of the dried distributed object is carried out, and the graphite-like matter which remained slightly is vanished nearly completely. Since the temperature to which a carbon nanotube disappears is higher than the graphite-like matter and amorphous carbon a little, it can vanish nearly completely the graphite-like matter and amorphous carbon which remain slightly, and can make only a carbon nanotube isolate by setting up burning temperature suitably. Therefore, as for burning temperature, it is preferably desirable to set it as 700-750 degrees C 500-800 degrees C. For 5 - 10 minutes is preferably suitable for a firing time for 1 - 30 minutes. Baking can be performed in air atmosphere.

[0011] Next, the example of this invention is described.

According to the conditions of a publication, the carbon nanotube rough product was obtained using the carbon cluster generation device type 3 made from an example vacuum metallurgy on the conditions of helium ** 500Torr, voltage 18V, and 8mm of diameters of an anode plate graphite in the above-mentioned reference. 0.253g of suspended matter which carried out ultrasonic irradiation of the 0.721g of this rough product for 60 minutes in ethanol after crushing with the mortar, and was produced was collected. 0.200g was supplied to the high-speed medium stirrer mill (content volume 100cm³) among those with ethanol 20cm³ and 200g (diameter of 1mm) of high toughness zirconia spheres, and it pulverized for 30 minutes at agitating-speed 3.6 m/s and the pulverization temperature of 8 degrees C. Next, it was made to distribute after drying the pulverizing object obtained by pulverizing, putting in and carrying out ultrasonic irradiation to 3 for 60 minutes 300cm of sodium-oleate 0.2wt% solution. At-long-intervals heart separation of this solution was carried out with the centrifugal force of 12000G for 30 minutes, and the supernatant containing the distributed object of 0.2 micrometers or less of pitch diameters was obtained. After carrying out the pressure filtration of this liquid using the filter of 0.45 micrometers of apertures, it pickled with 6 N-HNO₃ solution on the filter. Carbon nanotube 0.018g was obtained by calcinating 750 degrees C of samples on a filter for 5 minutes in air atmosphere. It checked that what was obtained was a carbon nanotube with the scanning-electron-microscope photograph (it is described as a SEM photograph below). Moreover, it was checked from this SEM photograph that a graphite particle and amorphous carbon are hardly contained.

[0012] In example of comparison 1 example 1, 750 degrees C of things which collected the suspended matter which carried out ultrasonic irradiation, and which was produced in ethanol, and were dried were calcinated for 5 minutes in air atmosphere without carrying-out operation-below pulverizing-processing-When this thing was observed with the SEM photograph, it remained in the state where the graphite particle about the diameter of 1 micrometer is not burned down, and it has not dissociated with a carbon nanotube.

[0013] In the example 1 of example of comparison 2 comparison, except having calcinated for 5 minutes at 800 degrees C, when what was obtained similarly was observed with the SEM photograph, the carbon nanotube had burned down and decreased compared with the thing of the example 1 of comparison.

[0014]

[Effect of the Invention] According to this invention, the quality carbon nanotube which does not contain a carbon impurity can be obtained by comparatively easy operation. Therefore, use in the very wide range field from a material science to electronics is expectable.

[Translation done.]